

Full counting statistics of nano-electromechanical systems

CHRISTIAN FLINDT¹(*), TOMÁŠ NOVOTNÝ^{1,2} and ANTTI-PEKKA JAUHO¹

¹ MIC – Department of Micro and Nanotechnology, Technical University of Denmark, DTU – Building 345east, DK-2800 Kongens Lyngby, Denmark

² Department of Electronic Structures, Faculty of Mathematics and Physics, Charles University – Ke Karlovu 5, 121 16 Prague, Czech Republic

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Abstract. – We develop a theory for the full counting statistics (FCS) for a class of nano-electromechanical systems (NEMS), describable by a Markovian generalized master equation. The theory is applied to two specific examples of current interest: vibrating C₆₀ molecules and quantum shuttles. We report a numerical evaluation of the first three cumulants for the C₆₀-setup; for the quantum shuttle we use the third cumulant to substantiate that the giant enhancement in noise observed at the shuttling transition is due to a slow switching between two competing conduction channels. Especially the last example illustrates the power of the FCS.

Introduction. – The full counting statistics (FCS) of charge transport in mesoscopic systems is an active topic of recent research [1–5]. Calculation and measurement of the whole probability distribution of transmitted charge is motivated by the fact that FCS provides more information about a particular system than just the mean current or current noise which are the first two cumulants of the large-time asymptotics of the probability distribution. Very recently, a measurement of the third cumulant, which quantifies the skewness of the distribution, was reported [6]. The detailed nature of charge transport in nanoelectromechanical systems (NEMS), another modern field in mesoscopics, poses many challenges both to experiments and theory, and the computation of FCS for NEMS is an important task that needs to be addressed. The first steps were taken recently with a calculation of FCS for a *driven, classical* shuttle [7].

In this Letter, we present a theory for the evaluation of cumulants in a wide class of NEMS encompassing the majority of systems considered thus far, namely those which can be described by a Markovian generalized master equation (GME). The current cumulants turn out to be fully determined by an extremal eigenvalue of the system evolution superoperator (Liouvillean) in analogy with previous studies [4, 5]. Their evaluation is, however, more complicated since in NEMS there are generally many relevant states which need to be taken into account. We solve the problem by formulating a systematic perturbation theory, and using this derive explicit formulas for the first three cumulants. The method is illustrated by

(*) E-mail: cf@mic.dtu.dk

two examples of NEMS – the C_{60} -experiment [8] and the quantum shuttle [9–11]. To test the method we calculate the first three cumulants for the model of the C_{60} -setup from [12]. In case the oscillator is equilibrated the cumulants can be calculated alternatively using $P(E)$ -theory which gives the same results. For the quantum shuttle we use the third cumulant to substantiate that the giant enhancement of the current noise in the transition region [10] is caused by a slowly fluctuating amplitude of the shuttle resulting in a slow switching between two current channels, *i.e.* tunneling and shuttling. This part complements [7], which considered a fixed driving amplitude, and [13] describing a related phenomenon in a different model.

Theory. – We consider a nanoelectromechanical system with discrete energy levels electronically coupled to two leads and mechanically coupled to a generic heat bath providing dissipation. The system is described by the reduced density operator $\hat{\rho}(t)$, which we assume evolves according to the Markovian GME

$$\dot{\hat{\rho}}(t) = \mathcal{L}\hat{\rho}(t). \quad (1)$$

The Liouvillian \mathcal{L} describes the dynamics of the system and we assume that the system tends exponentially to a stationary state $\hat{\rho}^{\text{stat}}$. This implies that the Liouvillian, which is a non-hermitian operator, has a single eigenvalue equal to zero with $\hat{\rho}^{\text{stat}}$ being the corresponding (normalized and unique) right eigenvector which we denote by $|0\rangle\rangle$ [16]. The corresponding left eigenvector is the identity operator $\hat{1}$, denoted by $\langle\langle 0|$, and we have $\langle\langle 0|0\rangle\rangle \equiv \text{Tr}(\hat{1}\hat{\rho}^{\text{stat}}) = 1$. The pair of eigenvectors allows us to define the projectors $\mathcal{P} \equiv |0\rangle\rangle\langle\langle 0|$ and $\mathcal{Q} \equiv 1 - \mathcal{P}$ obeying the relations $\mathcal{P}\mathcal{L} = \mathcal{L}\mathcal{P} = 0$ and $\mathcal{Q}\mathcal{L}\mathcal{Q} = \mathcal{L}$. We also introduce the pseudoinverse of the Liouvillian $\mathcal{R} \equiv \mathcal{Q}\mathcal{L}^{-1}\mathcal{Q}$, which is well-defined, since the inversion is performed only in the subspace spanned by \mathcal{Q} , where \mathcal{L} is regular. The assumption of exponential decay to the stationary state is equivalent to the spectrum of \mathcal{L} in the subspace spanned by \mathcal{Q} having a finite negative real part.

In order to evaluate the FCS of the system, *i.e.* the probability $P_n(t)$ of n electrons being collected in, say, the right lead in the time span t , we resolve the density operator $\hat{\rho}(t)$ and the GME with respect to n . The GME is a continuity equation for the probability (charge) and, therefore, we can identify terms corresponding to charge transfer processes between the system and the right lead. Specifically, we introduce the superoperator \mathcal{I}^+ of the particle current of electrons tunneling from the system to the right lead, and the corresponding superoperator \mathcal{I}^- of the reverse process, where electrons tunnel from the right lead to the system. In terms of these superoperators the n -resolved GME can be written as

$$\dot{\hat{\rho}}^{(n)}(t) = (\mathcal{L} - \mathcal{I}^+ - \mathcal{I}^-)\hat{\rho}^{(n)}(t) + \mathcal{I}^+\hat{\rho}^{(n-1)}(t) + \mathcal{I}^-\hat{\rho}^{(n+1)}(t) \quad (2)$$

with $n = \dots, -1, 0, 1, \dots$. From the n -resolved density operator we can obtain, at least in principle, the complete probability distribution $P_n(t) = \text{Tr}[\hat{\rho}^{(n)}(t)]$.

It is practical first to evaluate the cumulant generating function $S(t, \chi)$ defined as

$$e^{S(t, \chi)} = \sum_{n=-\infty}^{\infty} P_n(t) e^{in\chi}. \quad (3)$$

From $S(t, \chi)$ we then find the m 'th cumulant of the charge distribution (we take $e = 1$) by taking the m 'th derivative with respect to the counting field χ at $\chi = 0$, *i.e.* $\langle\langle n^m \rangle\rangle(t) = \frac{\partial^m S}{\partial (i\chi)^m} \big|_{\chi=0}$, and from the knowledge of all cumulants we can reconstruct $P_n(t)$. The cumulants of the current in the stationary limit $t \rightarrow \infty$ are given by the time derivative of the charge cumulants, *i.e.* $\langle\langle I^m \rangle\rangle = \frac{d}{dt} \langle\langle n^m \rangle\rangle(t) \big|_{t \rightarrow \infty}$. The first two current cumulants give the average current and the zero-frequency current noise, respectively.

Using $\hat{\rho}^{(n)}(t)$ we may express $S(t, \chi)$ as $e^{S(t, \chi)} = \text{Tr}[\sum_{n=-\infty}^{\infty} \hat{\rho}^{(n)}(t) e^{in\chi}] = \text{Tr}[\hat{F}(t, \chi)]$, where we have introduced the auxiliary operator $\hat{F}(t, \chi)$ whose equation of motion follows from the n -resolved GME,

$$\frac{\partial}{\partial t} \hat{F}(t, \chi) = [\mathcal{L} - (1 - e^{i\chi})\mathcal{I}^+ - (1 - e^{-i\chi})\mathcal{I}^-] \hat{F}(t, \chi) \equiv \mathcal{L}_\chi \hat{F}(t, \chi) \quad (4)$$

with the formal solution $\hat{F}(t, \chi) = e^{\mathcal{L}_\chi t} \hat{F}(0, \chi)$. We assume adiabatic evolution of the spectrum of \mathcal{L}_χ with increasing χ , *i.e.* there is a unique eigenvalue Λ_χ^{\min} of \mathcal{L}_χ associated with the projector \mathcal{P}_χ which develops from the zero eigenvalue of \mathcal{L} and which is the closest to zero for small enough χ . The rest of the spectrum still has finite negative real part which ensures the damping of its contribution for large times. Thus, we have

$$e^{S(t, \chi)} = \langle\langle \tilde{0} | e^{\mathcal{L}_\chi t} | F(0, \chi) \rangle\rangle \rightarrow e^{\Lambda_\chi^{\min} t} \langle\langle \tilde{0} | \mathcal{P}_\chi | F(0, \chi) \rangle\rangle = e^{\Lambda_\chi^{\min} t + C_\chi^{\text{init}}} \text{ for } t \rightarrow \infty, \quad (5)$$

where C_χ^{init} depends on the initial state of the system. However, the current cumulants in the stationary state do not depend on the initial conditions, but are totally determined by Λ_χ^{\min} in full analogy with previous studies [4, 5]. For NEMS in general \mathcal{L}_χ is of very large dimensions, and the numerical evaluation of higher order derivatives of Λ_χ^{\min} may become a formidable numerical problem. In order to circumvent this problem we determine Λ_χ^{\min} using Rayleigh-Schrödinger perturbation theory for $\mathcal{L}_\chi = \mathcal{L} + \mathcal{L}'_\chi$, treating \mathcal{L}'_χ as the perturbation. Since the Liouvillian is not hermitian, we cannot assume that it has a spectral decomposition in terms of its eigenvectors, and one cannot use directly the standard formulas. However, it is possible to formulate the perturbation theory exclusively in terms of the projectors \mathcal{P} , \mathcal{Q} and the pseudoinverse \mathcal{R} . As in standard Rayleigh-Schrödinger perturbation theory the first order correction is given by the average of the perturbation with respect to the unperturbed eigenstate. Taking the derivative of the first order correction with respect to $i\chi$ and letting $\chi \rightarrow 0$ we find

$$\langle\langle I \rangle\rangle = \langle\langle \tilde{0} | \mathcal{I} | 0 \rangle\rangle, \quad (6)$$

where $\mathcal{I} \equiv \mathcal{I}^+ - \mathcal{I}^-$. As expected the first cumulant equals the average current. For the second cumulant, *i.e.* the zero-frequency current noise, one finds

$$\langle\langle I^2 \rangle\rangle = \langle\langle \tilde{0} | \mathcal{J} | 0 \rangle\rangle - 2 \langle\langle \tilde{0} | \mathcal{I} \mathcal{R} \mathcal{I} | 0 \rangle\rangle, \quad (7)$$

where $\mathcal{J} \equiv \mathcal{I}^+ + \mathcal{I}^-$. In the high bias limit (where $\langle\langle \tilde{0} | \mathcal{I}^- | 0 \rangle\rangle = 0$, since backward tunneling is blocked) this expression yields the result previously derived in [16]. The expression for the third cumulant

$$\langle\langle I^3 \rangle\rangle = \langle\langle \tilde{0} | \mathcal{I} | 0 \rangle\rangle - 3 \langle\langle \tilde{0} | \mathcal{I} \mathcal{R} \mathcal{J} + \mathcal{J} \mathcal{R} \mathcal{I} | 0 \rangle\rangle - 6 \langle\langle \tilde{0} | \mathcal{I} \mathcal{R} (\mathcal{R} \mathcal{I} \mathcal{P} - \mathcal{I} \mathcal{R}) \mathcal{I} | 0 \rangle\rangle \quad (8)$$

is the main result of this section, and below we evaluate it for two specific cases. Higher order cumulants can be obtained in the same manner by calculating the corresponding higher order corrections.

Model 1: The C₆₀ experiment. – An experiment with a NEMS that has received much attention is the measurement of the *IV*-curves of a vibrating C₆₀-molecule [8]. The experiment has been modelled in several papers [12, 14, 15] using a model which will also be employed here. Calculations of *IV*-curves [12, 14] have been found to be in good agreement with the experiment, and the current noise has been predicted [15]. We calculate the third cumulant for this setup by applying our method to the model as described in [12].

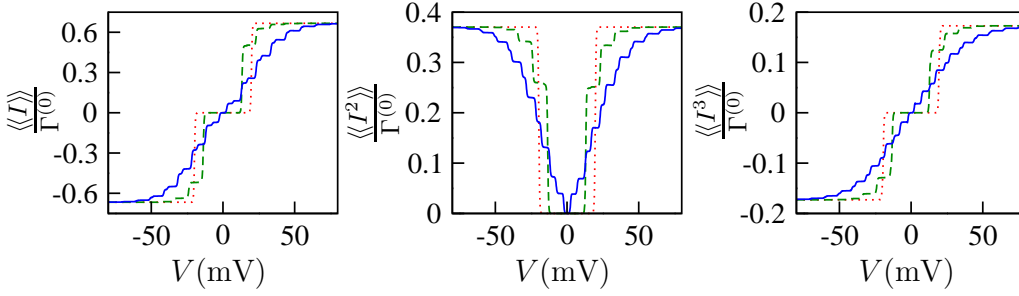


Fig. 1 – Results for the C_{60} -setup. First three cumulants as function of the bias V . The parameters, which correspond to fig. 3 of [12], are $E_c = 10$ meV, $\hbar\omega_0 = 5$ meV, $\hbar\Gamma^{(0)} = 1$ μ eV, $k_B T = 0.15$ meV, $K = 0.1\omega_0$, and $c_1 = 0$ (dashed), 0.8 (dotted), 1.5 (full), $c_2 = 0.005$.

In the model of [12] both the coupling to the leads and to the heat bath are treated in the weak coupling approximation which reduces the full GME to an ordinary Pauli master equation for the probabilities of occupation of the individual eigenstates of the system:

$$\begin{aligned} \frac{dP_{m,\sigma,l}}{dt} = & - \left[W_{l+1 \leftarrow l} + W_{l-1 \leftarrow l} + \sum_{m',\sigma',l',s} \Gamma_{m',\sigma',l' \leftarrow m,\sigma,l}^{(s)} \right] P_{m,\sigma,l} \\ & + \sum_{m',\sigma',l',s} \left[\Gamma_{m,\sigma,l \leftarrow m',\sigma',l'}^{(s)} + W_{l \leftarrow l'} (\delta_{l+1,l'} + \delta_{l-1,l'}) \delta_{\sigma,\sigma'} \delta_{m,m'} \right] P_{m',\sigma',l'}, \end{aligned} \quad (9)$$

where m, σ, l denote the (extra) charge on the molecule ($m = 0, 1$), the spin, and the vibrational state, respectively, while s indicates whether an electron tunnelled from/to the left ($s = -1$) or right ($s = 1$) lead. $P_{m,\sigma,l}$ is the probability of being in the eigenstate labelled by the subindices. Bath-mediated transitions between different vibrational states are given by the thermal rates

$$W_{l+1 \leftarrow l} = W_{l \leftarrow l+1} e^{-\hbar\omega_0/k_B T} = K \frac{l+1}{e^{\hbar\omega_0/k_B T} - 1}, \quad (10)$$

where ω_0 is the natural oscillator frequency. The charge transfer rates are

$$\begin{aligned} \Gamma_{1,\sigma,l' \leftarrow 0,0,l}^{(s)} &= \Gamma^{(0)} |\langle l' | e^{\gamma(\hat{a}^\dagger - \hat{a})} | l \rangle|^2 f(E_C + \frac{seV}{2} + \hbar\omega_0(l' - l - \gamma^2)), \\ \Gamma_{0,0,l \leftarrow 1,\sigma,l'}^{(s)} &= \Gamma^{(0)} |\langle l | e^{\gamma(\hat{a}^\dagger - \hat{a})} | l' \rangle|^2 [1 - f(E_C + \frac{seV}{2} + \hbar\omega_0(l' - l - \gamma^2))], \end{aligned} \quad (11)$$

where f is the Fermi function, $\Gamma^{(0)}$ is the bare tunneling rate, E_C is the charging energy difference, and V is the symmetrically applied bias. The quantity γ describes the bias-dependence of the electric field at the position of the molecule, and is assumed to have the form $\gamma = c_1 + \frac{eV}{\hbar\omega_0} c_2$ [12]. Here we do not consider the case where the rates depend on the position of the molecule, although this can easily be included.

The current superoperators are identified from the expression for the stationary current

$$I^{\text{stat}} = \underbrace{\sum_{\sigma,l,l'} \left[\Gamma_{0,0,l' \leftarrow 1,\sigma,l}^{(1)} P_{1,\sigma,l}^{\text{stat}} \right]}_{\langle\langle \hat{0} | \mathcal{I}^+ | 0 \rangle\rangle} - \underbrace{\sum_{\sigma,l,l'} \left[\Gamma_{1,\sigma,l' \leftarrow 0,0,l}^{(1)} P_{0,0,l}^{\text{stat}} \right]}_{\langle\langle \hat{0} | \mathcal{I}^- | 0 \rangle\rangle}. \quad (12)$$

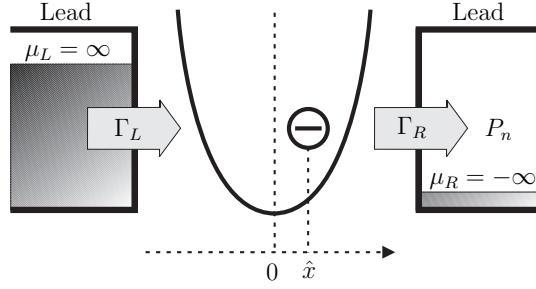


Fig. 2 – The quantum shuttle consists of a nanosized grain moving in a harmonic potential between two leads. A high bias between the leads drives electrons through the grain.

Here $|0\rangle\rangle$ is a diagonal density matrix containing the stationary probabilities $P_{m,\sigma,l}^{\text{stat}}$. Due to this diagonal form of the density matrices the relevant superoperators needed for the cumulants can be represented by matrices of dimension $2N \times 2N$ (N is the number of vibrational modes) which makes the calculation of the cumulants numerically straightforward.

In fig. 1 we show the bias-dependence of the first three cumulants for parameters corresponding to fig. 3 of [12]. Since $\Gamma^{(0)} \ll K$, the oscillator is in equilibrium and, therefore, the FCS of the model can also be calculated from a simple two-level model with 4 rates given by $P(E)$ -theory. We have verified that the semi-analytical results coincide with numerics (not shown) which we view as a non-trivial test of our method. In non-equilibrium cases there are no simple alternatives to the numerics. We demonstrate the full power of the method in the second example.

Model 2: The Quantum Shuttle. – We consider the model of a quantum shuttle used in [9–11]. The system consists of an oscillating nanoscopic grain coupled to two leads (fig. 2). In the strong Coulomb blockade regime the grain effectively has just one electronic level. The oscillations of the grain are treated fully quantum mechanically, and damping of the oscillations is due to a surrounding heat bath. As in [10] we consider the n -resolved system density matrices $\hat{\rho}_{ii}^{(n)}(t)$, $i = 0, 1$, where n is the number of electrons that have tunneled into the right lead in the time span t . In the high bias limit n is nonnegative, and the $\hat{\rho}_{ii}^{(n)}(t)$ evolve according to the n -resolved GME

$$\begin{aligned} \dot{\hat{\rho}}_{00}^{(n)}(t) &= \frac{1}{i\hbar} [\hat{H}_{\text{osc}}, \hat{\rho}_{00}^{(n)}(t)] + \mathcal{L}_{\text{damp}} \hat{\rho}_{00}^{(n)}(t) - \frac{\Gamma_L}{2} \{e^{-\frac{2\hat{x}}{\lambda}}, \hat{\rho}_{00}^{(n)}(t)\} + \Gamma_R e^{\frac{\hat{x}}{\lambda}} \hat{\rho}_{11}^{(n-1)}(t) e^{-\frac{\hat{x}}{\lambda}}, \\ \dot{\hat{\rho}}_{11}^{(n)}(t) &= \frac{1}{i\hbar} [\hat{H}_{\text{osc}} - eE\hat{x}, \hat{\rho}_{11}^{(n)}(t)] + \mathcal{L}_{\text{damp}} \hat{\rho}_{11}^{(n)}(t) - \frac{\Gamma_R}{2} \{e^{\frac{2\hat{x}}{\lambda}}, \hat{\rho}_{11}^{(n)}(t)\} + \Gamma_L e^{-\frac{\hat{x}}{\lambda}} \hat{\rho}_{00}^{(n)}(t) e^{-\frac{\hat{x}}{\lambda}}, \end{aligned} \quad (13)$$

with $n = 0, 1, \dots$ and $\hat{\rho}_{11}^{(-1)}(t) \equiv 0$. Here the commutators describe the coherent evolution of the charged (ρ_{11}) or empty (ρ_{00}) shuttle which is modelled by a quantum mechanical harmonic oscillator of mass m and frequency ω . The electric field between the leads is denoted E . The terms proportional to $\Gamma_{L/R}$ describe transfer processes from the left/to the right lead with hopping amplitudes that depend exponentially on the position $\frac{\hat{x}}{\lambda}$, where λ is the electron tunneling length. The mechanical damping of the oscillator is described by the damping kernel (here $T = 0$) $\mathcal{L}_{\text{damp}} \hat{\rho} = -\frac{i\gamma}{2\hbar} [\hat{x}, \{\hat{\rho}, \hat{\rho}\}] - \frac{\gamma m \omega}{2\hbar} [\hat{x}, [\hat{x}, \hat{\rho}]]$ [9, 10]. We identify the current superoperators from (13): $\mathcal{I}^+ \hat{\rho} = \Gamma_R e^{\frac{\hat{x}}{\lambda}} |0\rangle\langle 1| \hat{\rho} |1\rangle\langle 0| e^{-\frac{\hat{x}}{\lambda}}$, $\mathcal{I}^- \equiv 0$.

In [9, 10] it was found that the quantum shuttle exhibits a crossover from tunneling to

shuttling when the damping, starting above a certain threshold value, is decreased. This transition is clearly recorded both in the current [9] and the zero-frequency current noise [10]. The FCS in the tunneling and shuttling limit is to a first approximation captured by the results for the zero amplitude (with appropriately renormalized rates) and the large (shuttling) amplitude of a driven shuttle [7], respectively. When approaching the semi-classical regime, a giant enhancement of the noise was found in the transition region. This behavior was tentatively attributed to amplitude fluctuations in the spirit of [13], however, a more quantitative explanation has been missing. The phase space representation of the stationary state of the shuttle in the transition region indicated that shuttling and tunneling processes coexist [10] leading to the conjecture that the giant noise enhancement is caused by switching between two current channels⁽¹⁾ (tunneling and shuttling) induced by infrequent jumps between two discrete values of the shuttle amplitude. Very recently the FCS of such bistable systems has been studied [18], and it was found that the first three cumulants are (assuming that the individual channels are noiseless)

$$\langle\langle I \rangle\rangle = \frac{I_S \Gamma_{S \leftarrow T} + I_T \Gamma_{T \leftarrow S}}{\Gamma_{T \leftarrow S} + \Gamma_{S \leftarrow T}}, \quad (14a)$$

$$\langle\langle I^2 \rangle\rangle = 2(I_S - I_T)^2 \frac{\Gamma_{S \leftarrow T} \Gamma_{T \leftarrow S}}{(\Gamma_{S \leftarrow T} + \Gamma_{T \leftarrow S})^3}, \quad (14b)$$

$$\langle\langle I^3 \rangle\rangle = 6(I_S - I_T)^3 \frac{\Gamma_{S \leftarrow T} \Gamma_{T \leftarrow S} (\Gamma_{T \leftarrow S} - \Gamma_{S \leftarrow T})}{(\Gamma_{S \leftarrow T} + \Gamma_{T \leftarrow S})^5}. \quad (14c)$$

Here $I_{S/T}$ denote the current associated with the shuttling/tunneling channel⁽²⁾, while $\Gamma_{T \leftarrow S}$ is the transition rate from the shuttling to the tunneling channel and $\Gamma_{S \leftarrow T}$ is the rate of the reverse transition.

The rates can be evaluated analytically in the limit $\lambda \rightarrow \infty$ and $E \rightarrow 0$ [19]. However, for other parameters an alternative approach is needed. First, one evaluates the first three cumulants numerically following the theory presented above. Then, one calculates the rates from the first two cumulants (eqs. (14a,14b)) and compares the numerically calculated third cumulant from eq. (8) with the one obtained from eq. (14c). The numerical calculation of the cumulants is only possible by a truncation of the oscillator Hilbert space. By retaining the N lowest oscillator states the (non-sparse) matrix representations of the superoperators entering eqs. (6-8) are of dimension $2N^2 \times 2N^2$, which for the required values of $N \sim 100$ leaves us with non-trivial numerical matrix problems. However, using the iterative methods described in [16] the cumulants can be evaluated numerically.

If fig. 3 we show the γ -dependence of the first three cumulants for $\lambda = 1.5x_0$, where $x_0 = \sqrt{\hbar/m\omega}$. The first cumulant, the current, shows the transition from the tunneling to shuttling current with decreasing damping. The transition is also evident from the second cumulant, the zero-frequency current noise, which exhibits a giant enhancement in the transition region, before dropping to very low values in the shuttling region. Together with the numerical results for the third cumulant we show the analytic expression (eq. (14c)) with rates extracted from the first two cumulants. As can be seen the two data sets coincide, which we take as evidence that the quantum shuttle in the transition region indeed behaves as a bistable system for which the FCS is known [18]. When approaching the deep quantum regime, $\lambda \sim x_0$ (not shown), the transition from tunneling to shuttling is smeared out and the distinct current channels cease to exist. In this limit the bistable system model is not valid.

⁽¹⁾Such a behavior, referred to as the ‘whistle’ effect, was first reported in [17].

⁽²⁾ $I_S = \omega/2\pi$, $I_T = \frac{\tilde{\Gamma}_L \tilde{\Gamma}_R}{\tilde{\Gamma}_L + \tilde{\Gamma}_R}$, with $\tilde{\Gamma}_R = \Gamma_R e^{\hbar/m\omega\lambda^2} e^{2eE/m\omega^2\lambda}$, $\tilde{\Gamma}_L = \Gamma_L e^{\hbar/m\omega\lambda^2}$ [10, 19].

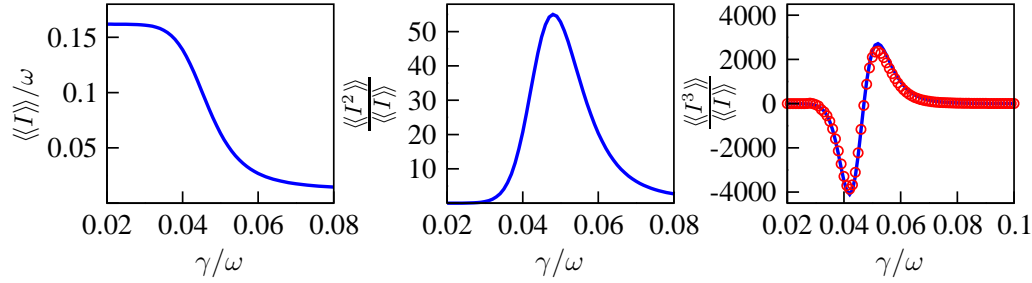


Fig. 3 – Results for the quantum shuttle. First three cumulants as function of the damping γ . The parameters are $\Gamma_L = \Gamma_R = 0.01\omega$, $\lambda = 1.5x_0$, $d \equiv eE/m\omega^2 = 0.5x_0$, $x_0 = \sqrt{\hbar/m\omega}$. The full lines indicate numerical results, while the circles indicate the analytic expression for the third cumulant assuming that the shuttle in the transition region effectively behaves as a bistable system.

Conclusion. – We have presented a method for computation of the FCS for typical nanoelectromechanical systems and applied it to two specific models. For the C_{60} -setup with equilibrated oscillator we have calculated the first three cumulants and explained the results in terms of a simple two-level model. For the quantum shuttle we have used the first three cumulants as evidence that the shuttle in the transition region behaves as a bistable system. This example clearly illustrates the usefulness of the FCS in probing a microscopic system. Here we have only shown explicit expressions for the first three cumulants, our method, however, can be extended to the calculation of cumulants of any order, and we believe that the method has a broad range of applicability.

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